



PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of

Hiroyuki MOCHIZUKI et al.

Group Art Unit: 1794

Application No.: 10/572,643

Examiner: B. CROUSE

Filed: March 20, 2006

Docket No.: 127380

For: ORGANIC ELECTROLUMINESCENT ELEMENT AND MANUFACTURING
METHOD THEREOF

DECLARATION UNDER 37 C.F.R. §1.132

I, Hiroyuki MOCHIZUKI, a citizen of Japan, hereby declare and state:

1. I received a doctoral degree from the Interdisciplinary Graduate School of Science and Engineering, Tokyo Institute of Technology, in March 2001, where my doctoral thesis was in the field of organic electroluminescent devices.

2. I have been employed by the National Institute of Advanced Industrial Science and Technology (AIST) from April, 2001 to March, 2009 and I have a total of more than eight years of work and research experience in polymer opto-electronic devices. Further, I have been employed by New Energy and Industrial Technology Development Organization (NEDO) since April, 2009 and I have a total of more than seven additional months of work and research experience in polymer opto-electronic devices.

3. I am a named inventor in the above-identified patent application.

4. I have a professional relationship, as an employee, with an assignee of the above-identified patent application. In the course of that professional relationship, I received compensation directly from the assignee for my work relating to polymer opto-electronic

devices. I am not being compensated for my work in connection with this Declaration, other than my regular compensation as an employee.

5. I and/or those under my direct supervision and control have conducted the following experimental tests.

Five electroluminescent devices were prepared: (1) Example A; (2) Comparative Example A1; (3) Comparative Example A2; (4) Comparative Example A3; and (5) Comparative Example A4. Example A was prepared using the method according to Example 1 recited in the specification (see specification, pages 11-12); Comparative Example A1 was prepared using a conventional spin coating method; Comparative Example A2 was prepared using only the contact and penetration method recited in the specification (see specification, page 11); and Comparative Examples A3 and A4 were prepared using a vapor deposition method.

With respect to preparing Comparative Example A3 using a vapor deposition method, a resin thin film (1 mm thick, 8 mm wide, 40 mm long) of PPV formed on a glass substrate, which had an ITO electrode was inserted into a vacuum deposition apparatus. An electron transport compound 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole (PBD) was deposited on the PPV surface in thicknesses of 35 nm and 70 nm, using a deposition speed of 0.2 nm/second. Subsequently, silver and magnesium were deposited together to laminate a negative electrode, thereby producing the electroluminescent elements (see Fig. 1, below).

With respect to preparing Comparative Example A4 using a vapor deposition method, Comparative Example A4 was prepared in a similar manner to Comparative Example A3, except that prior to laminating the negative electrode, cotton absorbed with acetone was placed in contact with the PBD surfaces of these samples. The PBD dissolved as a result of contact with the acetone, and, thus, the PBD was wiped off the surface of these samples. Because the PBD dissolved upon contact with acetone, the PPV surface of the

samples had the same appearance as they did prior to depositing the PBD (see Fig. 2, below).

After the acetone-wiped PPV surface was dried, silver and magnesium were deposited together to laminate a negative electrode, thereby producing the electroluminescent element (see Fig. 3, below). This electroluminescent element of Comparative Example A4 emitted yellowish green light, and had a maximum luminance of 19 cd/m² at 14V. The external quantum efficiency was 0.71 lm/w.

Furthermore, when the PPV surface on which PBD was deposited in the above-mentioned preparation was heated for two minutes at 110°C, condensation was observed on the surface. When the acetone-absorbed cotton was put into contact with the PBD surface of these samples, PBD was wiped off the surface of these samples. After wiping, these samples had the same PPV surface as existed prior to depositing PBD (see Fig. 2, below).

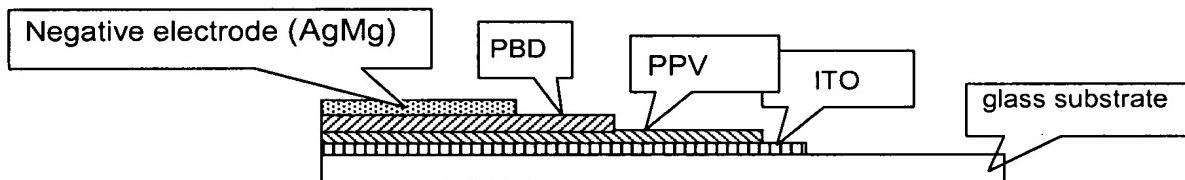


Fig. 1

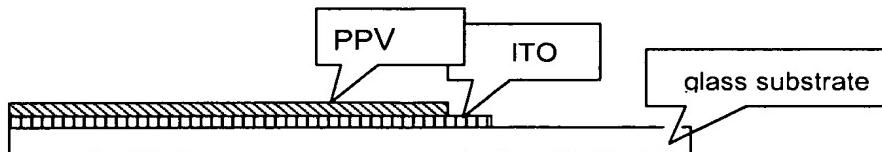


Fig. 2

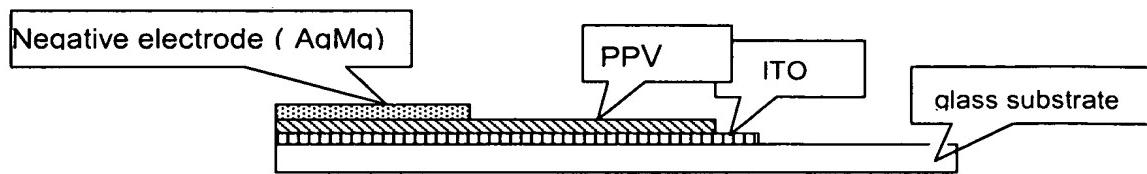


Fig. 3

Tables summarizing the methodology and the results are provided below:

Table 1
Doping of an Unsubstituted π Conjugated Organic Compound with a Charge Transport Material
Optical Characteristics of Electroluminescent Devices

Electroluminescent Device	Methodology	Observations and Results	Maximum Luminance	Quantum Efficiency
Example A Applicants' Specification As recited in Example 1 of Applicants' specification (see pages 11, line 7 to page 12, line 3)	1. Electroluminescent device emitted a yellowish green light 2. Quantity of PBD that had penetrated PPV was 2.7 wt. % (15,689.5% increase from A4)	3000 cd/m ² at 14V (350.7% increase from A4)	3.2 lm/w	
Comparative Example A1 Traditional Spin coating method (See e.g. Yu, col. 5, line 64 - col. 6, line 49)	1. When PPV and 2.7 wt. % PBD were placed in 100 ml of chloroform and stirred for 24 hours, the PBD dissolved while the PPV remained completely insoluble. 2. With this mixed liquid, film formation was impossible to attain using inkjet methods, blade coating methods and/or screen printing methods. 3. Thus, an electroluminescent element could not be produced.	<i>See "Observations and Results"</i>		

Electroluminescent Device	Methodology	Observations and Results <i>See "Observations and Results"</i>	Maximum Luminance Quantum Efficiency <i>See "Observations and Results"</i>
Comparative Example A2	Contact and Penetration Method PPV applied on a glass substrate	<ol style="list-style-type: none"> 1. PPV applied on a glass substrate cannot solvate in water or an organic solvent. 2. Although poly(p-xylene thiophenium chloride) as a precursor of PPV can solvate in water, PBD cannot solvate in poly(p-xylene thiophenium chloride) solution. 3. In addition, PBD is not able to solvate in a solution in which poly(p-xylene thiophenium chloride) is solvated in methanol. 	
Comparative Example A3		Vapor Deposition Method <i>(See e.g. Seo, paragraph [0033]; Matsuo, paragraph [0248]; Tang, col. 11, line 50 to col. 12, line 13).</i>	<ol style="list-style-type: none"> 1. After PBD was deposited on PPV surfaces having thicknesses of 35 nm and 70 nm in PBD, although Direct-current voltage was applied to each of these electroluminescent elements with 35 nm in thickness and 70 nm in thickness of PBD, neither of the electroluminescent elements could emit light.

Electroluminescent Device	Methodology	Observations and Results	Maximum Luminance	Quantum Efficiency
Comparative Example A4	Vapor Deposition Method <i>(See e.g. Seo, paragraph [0033]; Matsuo, paragraph [0248]; Tang, col. 11, line 50 to col. 12, line 13).</i>	<p>1. The PBD dissolved as a result of the contact with acetone, and, thus, PBD was wiped off the surface of these samples. Because the PBD dissolved upon contact with acetone, the samples had the same appearance as the PPV surface prior to depositing the PBD.</p> <p>2. After the acetone-wiped PPV surface was dried, silver and magnesium were deposited together to laminate a negative electrode, thereby producing the electroluminescent element.</p> <p>3. Also, when the PPV surface on which PPB was deposited in the above-mentioned preparation was heated for two minutes at 110°C, condensation was observed on the surface. When the cotton containing acetone was put into contact with the PBD surface of these samples, PBD was wiped off the surface of these samples. After wiping, these samples had the same PPV surface as existed prior to depositing PBD.</p>	19 cd/m ² at 14V (99.4% decrease from Example A)	0.71 lm/w (77.8% decrease from Example A)

In the development of a display monitor, the luminance must be approximately 1000 cd/m² or greater, although it may vary depending on the fineness of the pixels. If the luminance is less than 1000 cd/m², it may not be possible to recognize an image in a typical room environment (under fluorescent light). If the efficiency is 1 lm/w or less, then power consumption is large, normal batteries are consumed in just several minutes of lighting, and the heating value is so high that the element itself may be damaged. The unit "lm/w" used herein denotes "lumen/watt."

As shown in Table 1, when manufacturing an organic electroluminescent element using the unsubstituted π conjugated organic polymer compound PPV, the organic electroluminescent element cannot be produced using: (1) a traditional spin coating method (Comparative Example A1); or (2) a contact and penetration method (Comparative Example A2). With respect to the samples prepared using a vapor deposition method (Comparative Examples A3 and A4), it was observed that even if PBD is deposited on PPV under vapor deposition, PBD does not penetrate into the PPV layer, and the performance of the electroluminescent element deteriorates remarkably. Further, the PBD which was wiped off after coming into contact with the acetone-absorbed cotton did not affect the efficiency of the electroluminescent device since it failed earlier to permeate into the PPV layer.

Compared to using a vapor deposition method, the method of the present application resulted in a **15,698.5% increase** in the measured maximum luminance and a **350.7% increase** in quantum efficiency over the vapor deposition method. These sizeable percent increases in the maximum luminance and quantum efficiency between electroluminescent devices prepared by the method of Example 1 and a vapor deposition method were vastly improved and unanticipated.

I hereby declare that all statements made herein of my own knowledge are true, and that all statements made on information and belief are believed to be true; and

further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine and/or imprisonment under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing therefrom.

Date: Nov. 17 2009

Hiroyuki Mochizuki
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